

Laser-Induced Material Dynamics

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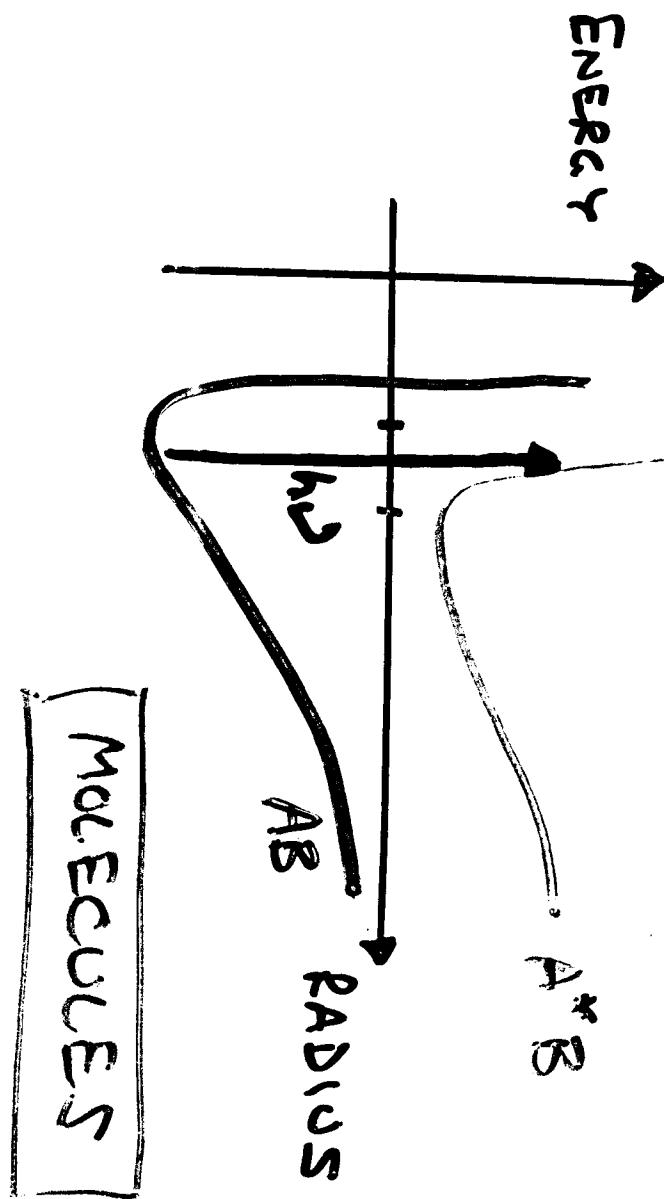
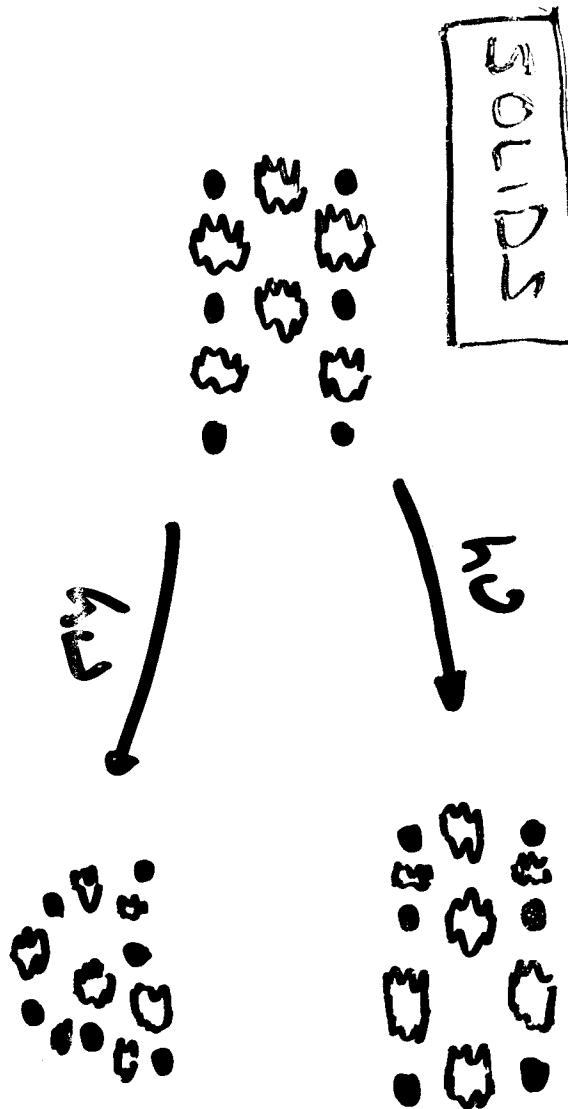
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Overview

- **Motivation:** use time-resolved x-ray diffraction as a real-time probe of atomic motions to study material dynamics.
- **Apparatus:** Beamline: focused, monochromated, intermediate x-rays; synchronized ultrashort-pulse laser; ps-resolution x-ray detector.
- **Results :** observation of laser-induced coherent acoustic phonons and non-thermal melting.

- = ATOMS
- \curvearrowleft = ELECTRONS

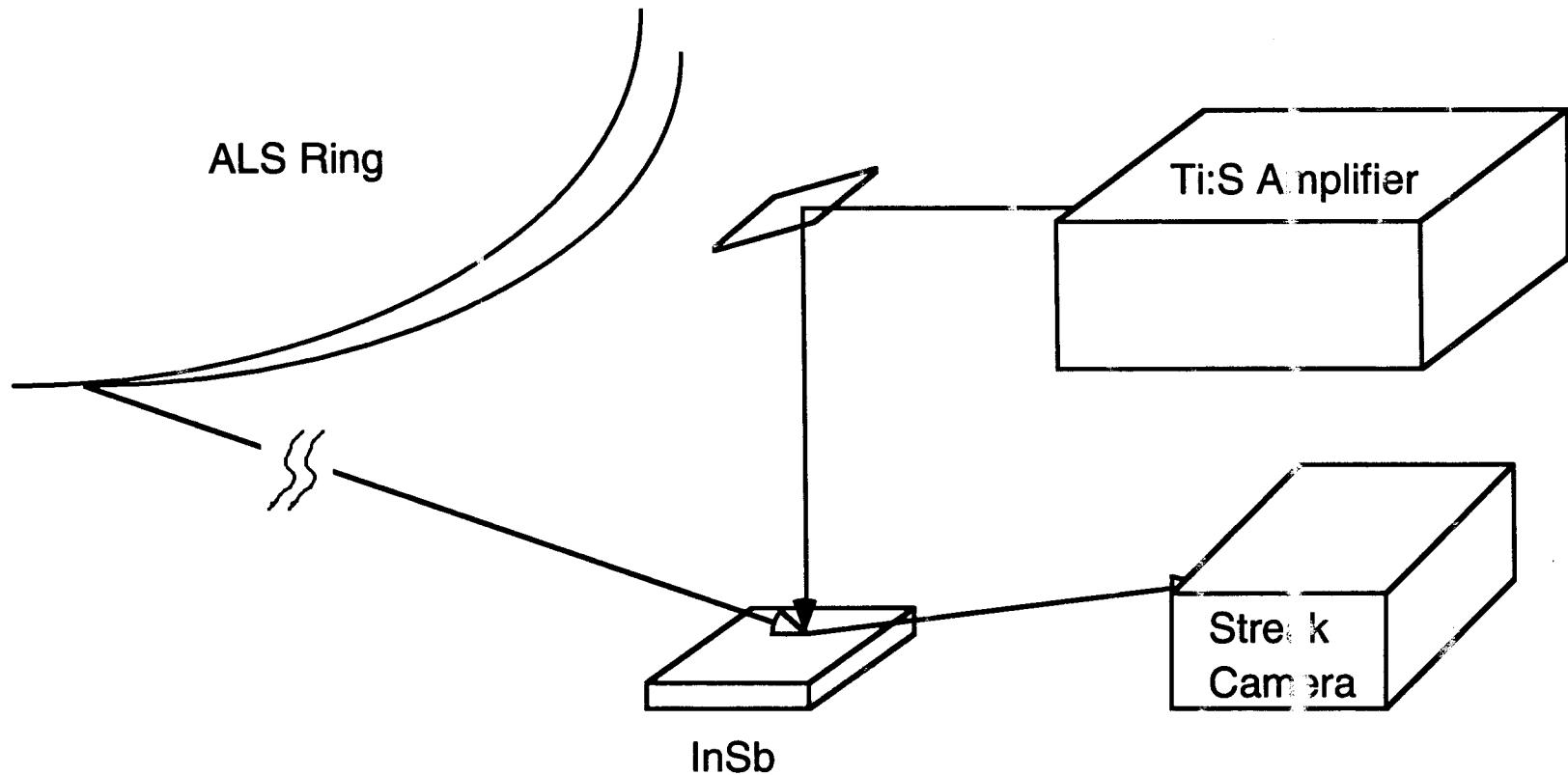


At a laser intensity corresponding to
absorption of about one photon per atom,
what happens to a solid?

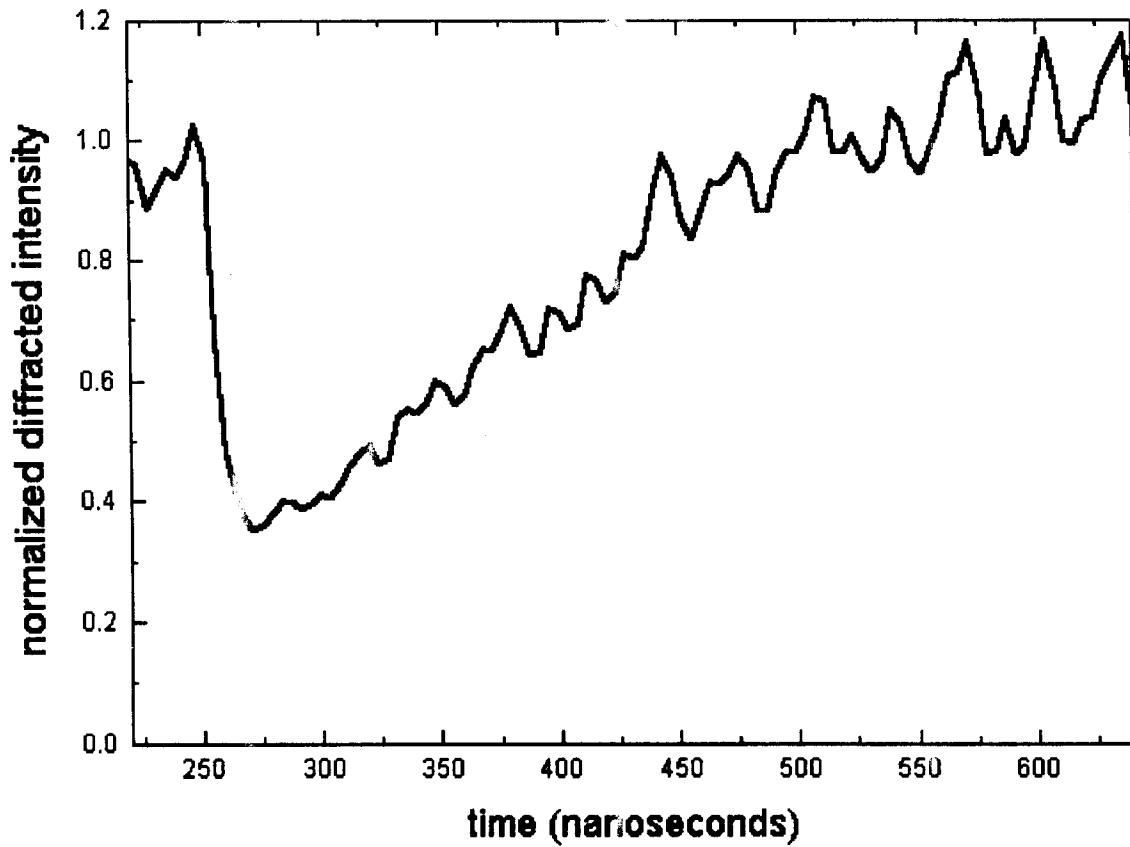
$$T_{\text{pulse}} < \frac{\text{atomic spacing}}{\text{sound velocity}} = \frac{10^{-8} \text{ cm}}{10^5 \text{ cm/s}}$$

$T_{\text{pulse}} < 10^{-13} \text{ s}$

Experimental Setup

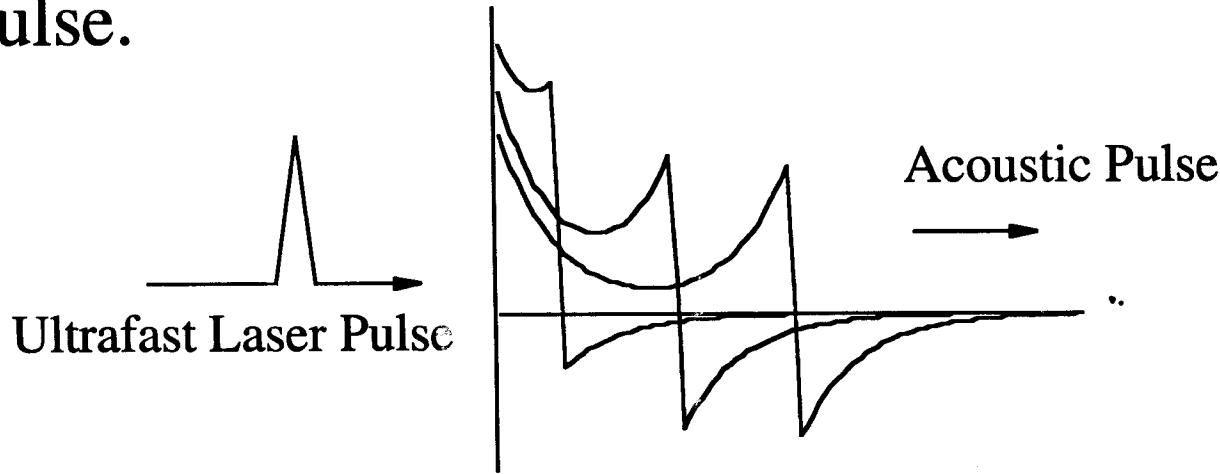


Time-resolved Bragg intensity from laser-heated InSb (long-time scan)



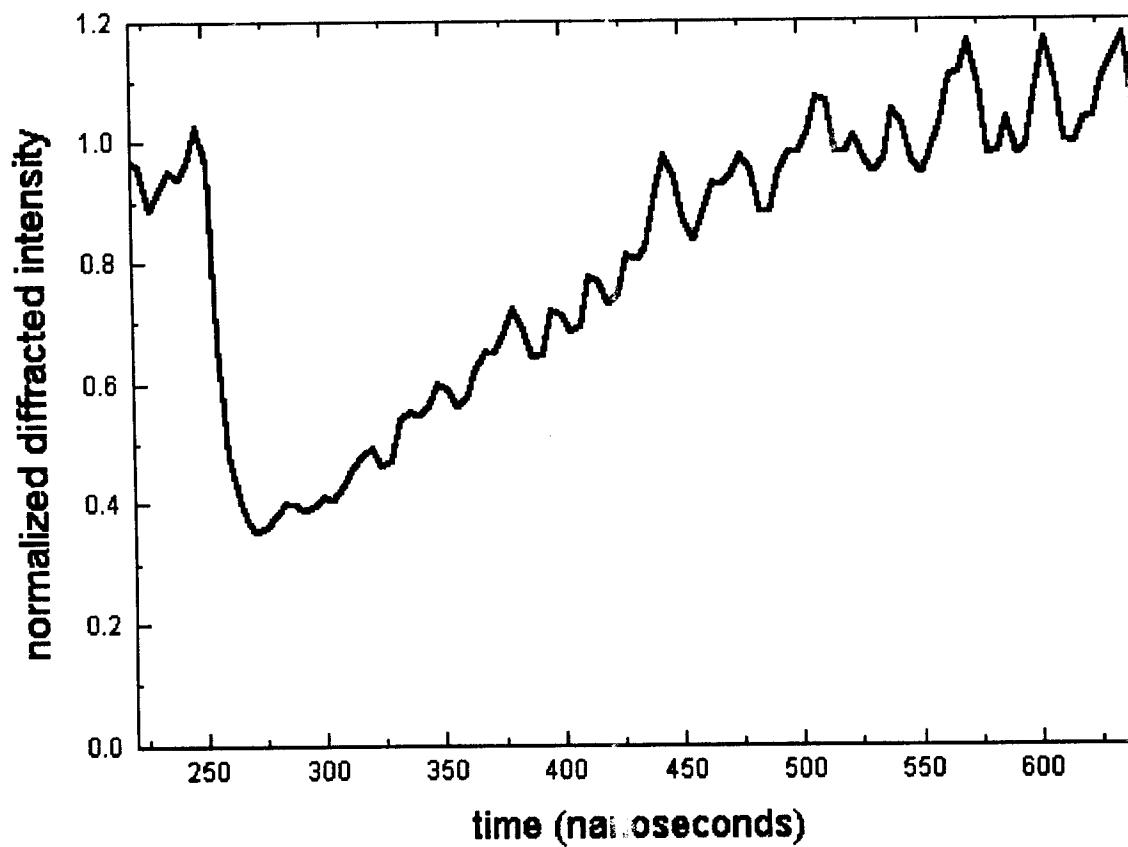
Generation of Coherent Acoustic Phonons

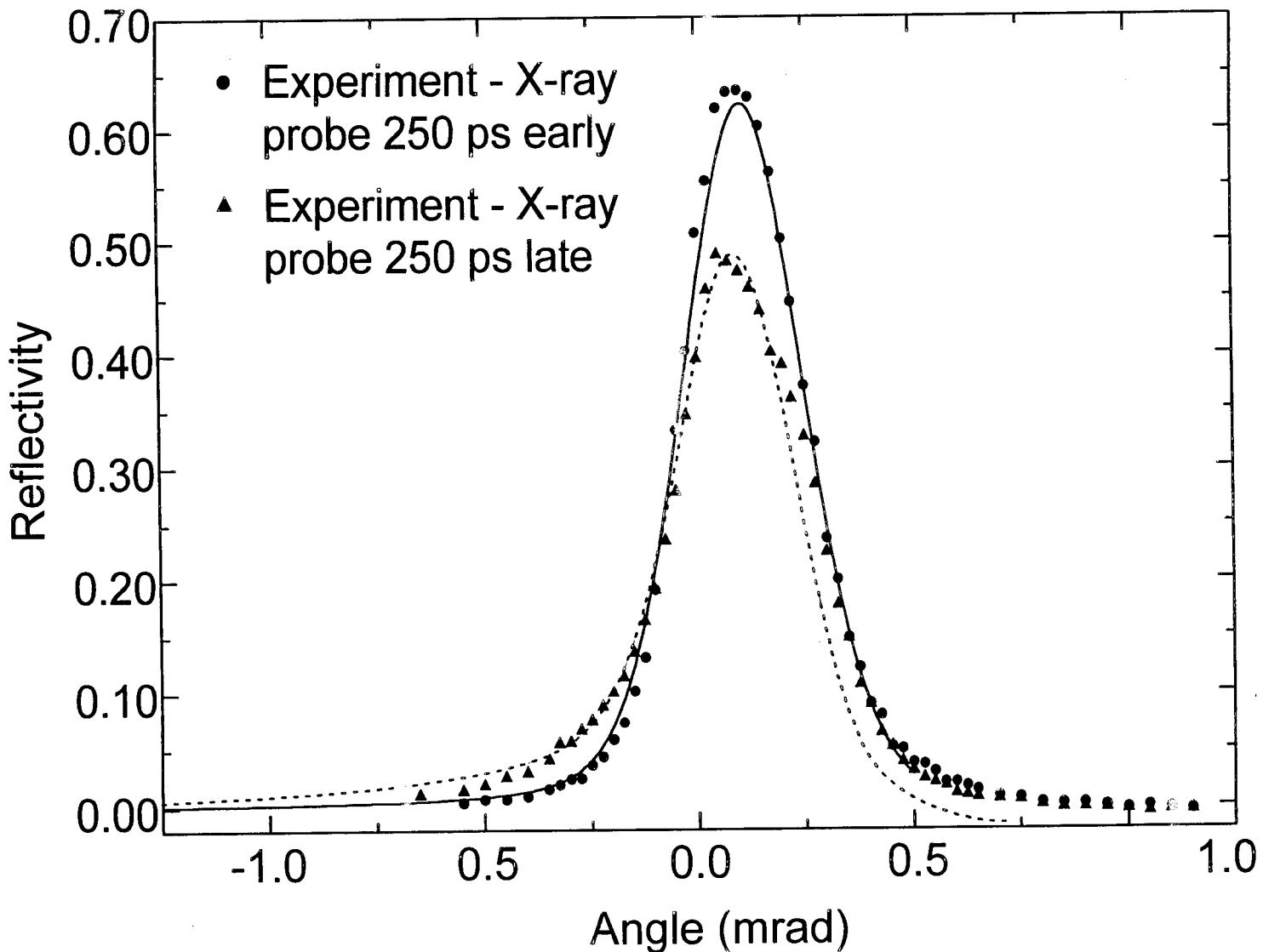
- Ultrafast laser pulse launches coherent acoustic pulse.



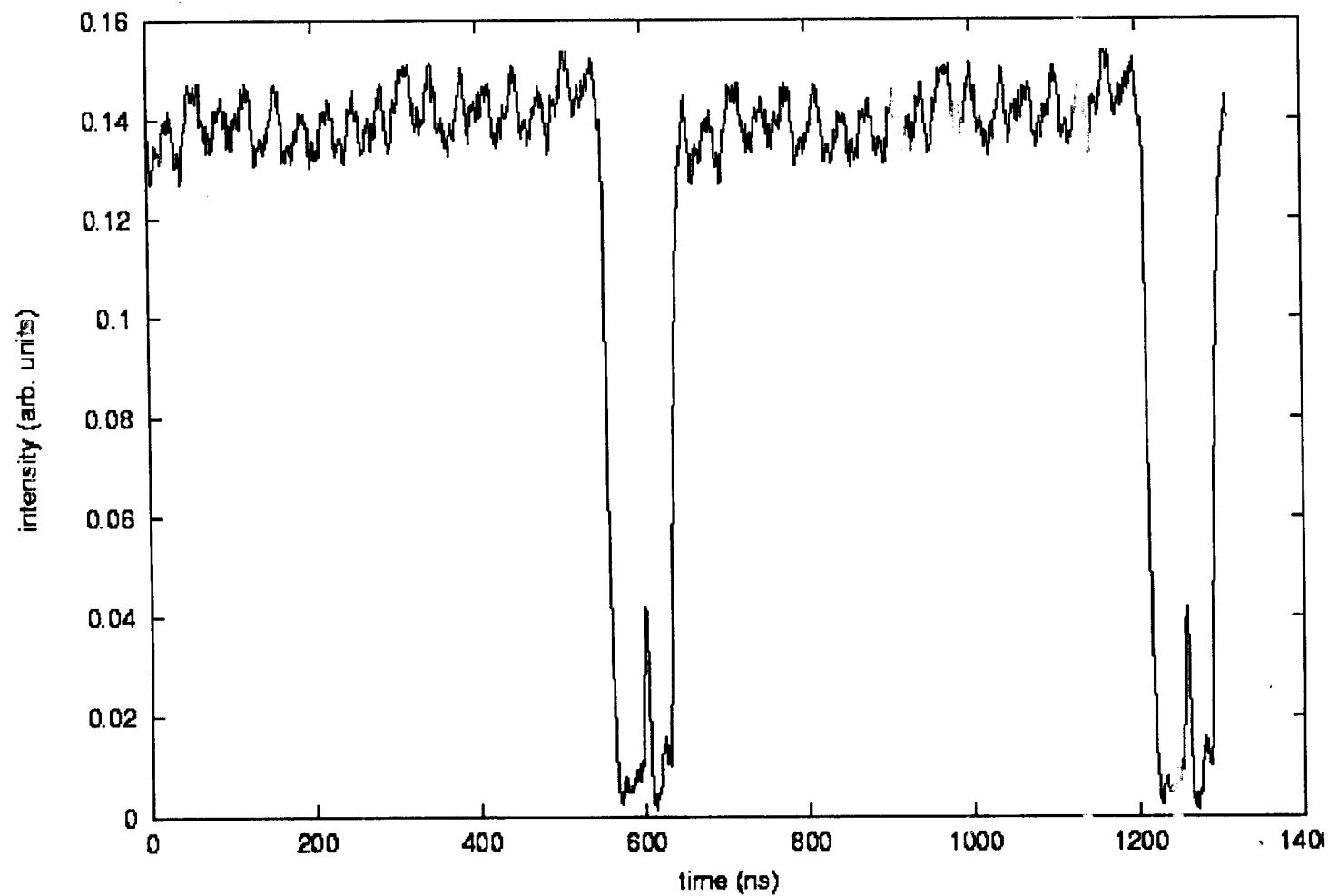
- Coherent acoustic pulse = coherent superposition of acoustic phonons with $\Delta q \sim 1/(\text{laser penetration depth})$.

Time-resolved Bragg intensity from laser-heated InSb (long-time scan)





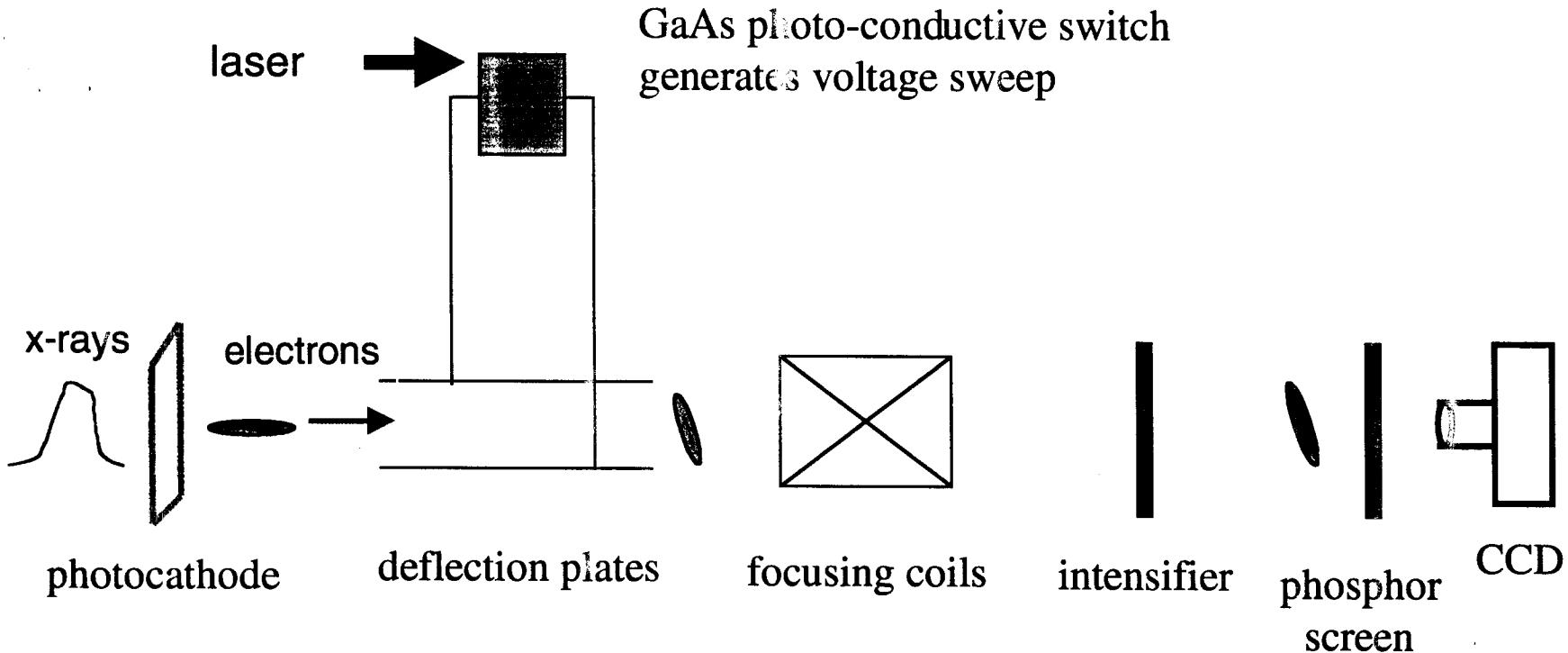
ALS Pulse Timing



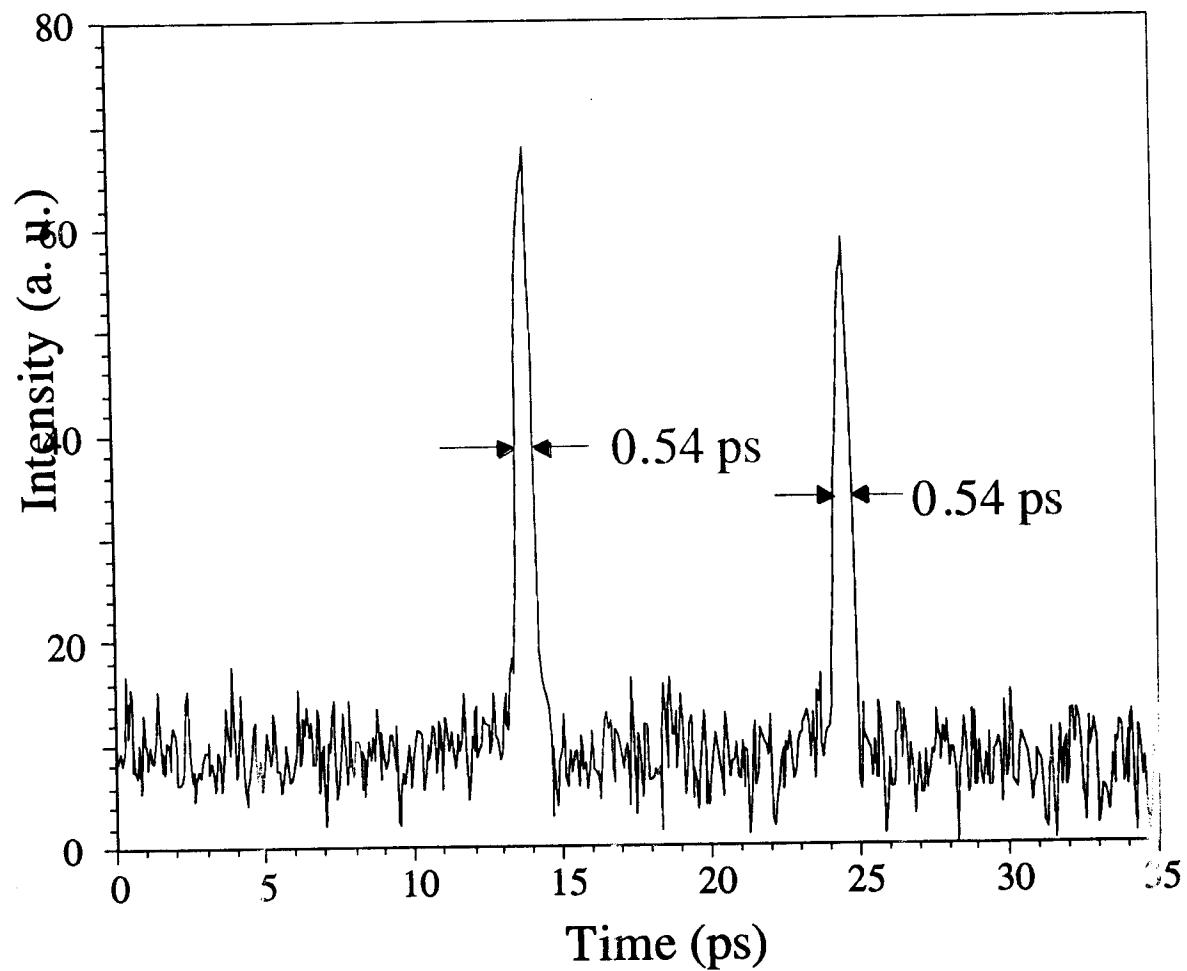
Synchronization of a 100 fs Ti:Al₂O₃ laser to the ALS synchrotron radiation source enables x-ray diffraction studies on three different timescales

- 1) Recording of transients; Detector limited; APD ~10 ns
- 2) Pump/probe ; the ALS pulse duration ; ~70 ps
- 3) Ultrafast; Cross-correlation or Streakcamera ; <2 ps

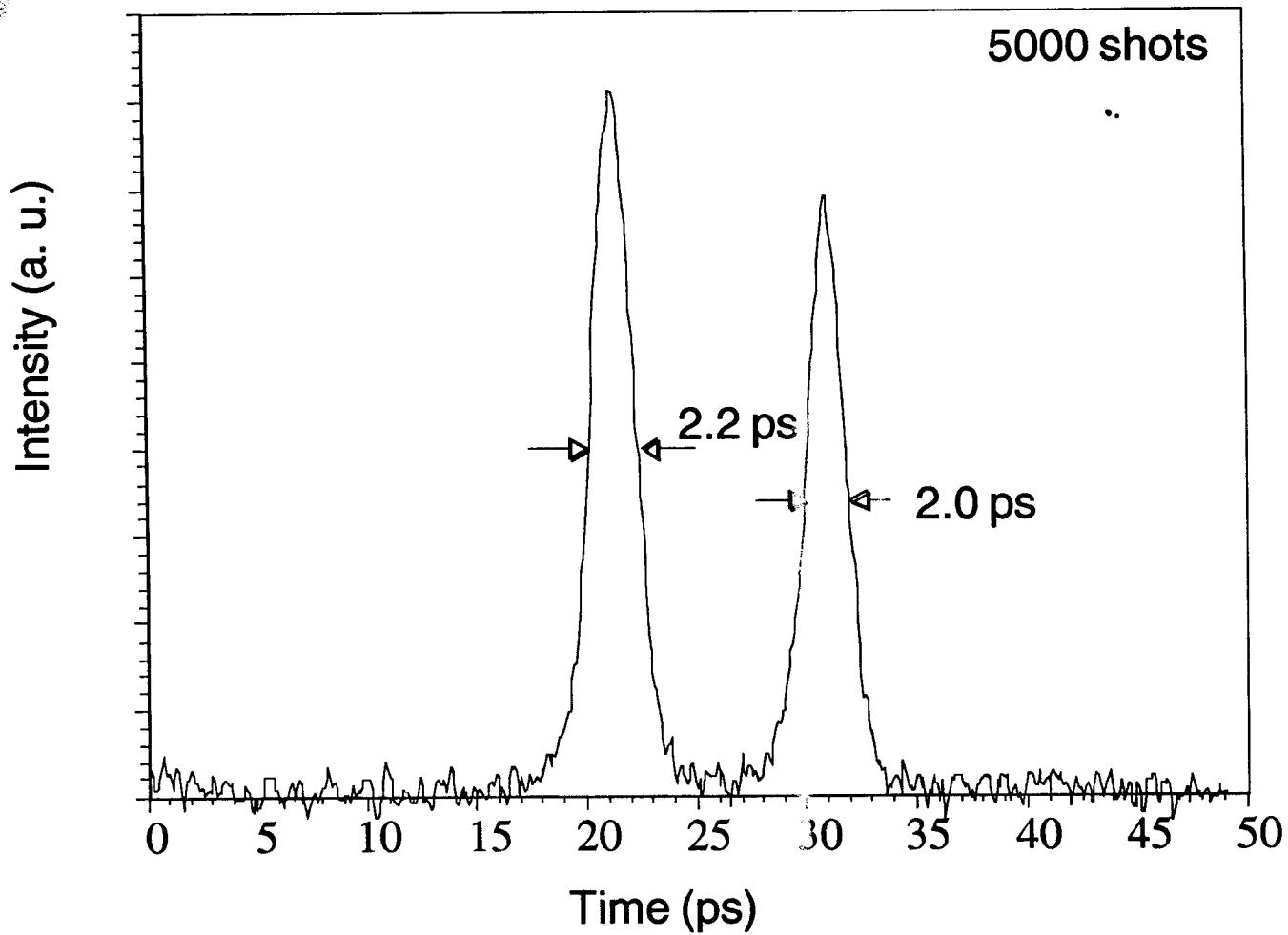
Picosecond x-ray streak camera detector

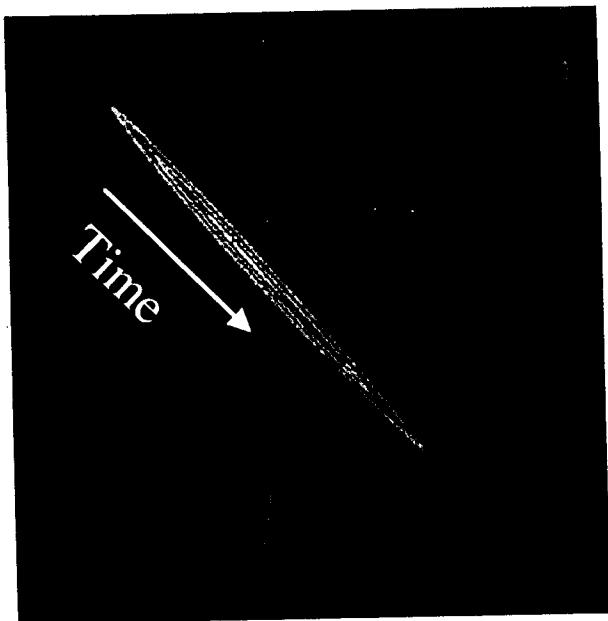


Single Shot Resolution

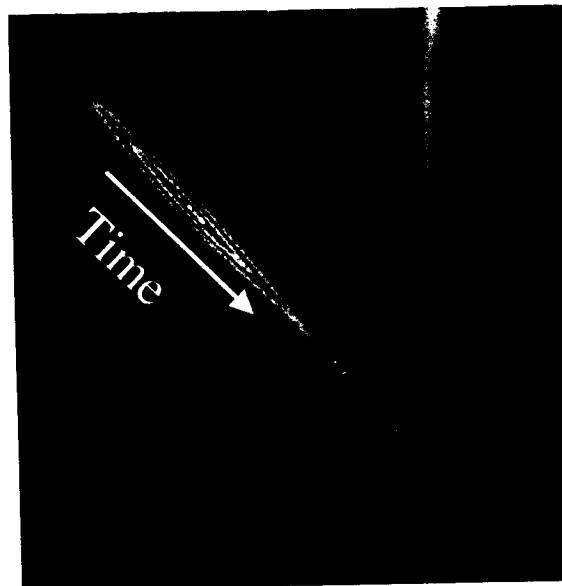


Jitter Limited Resolution





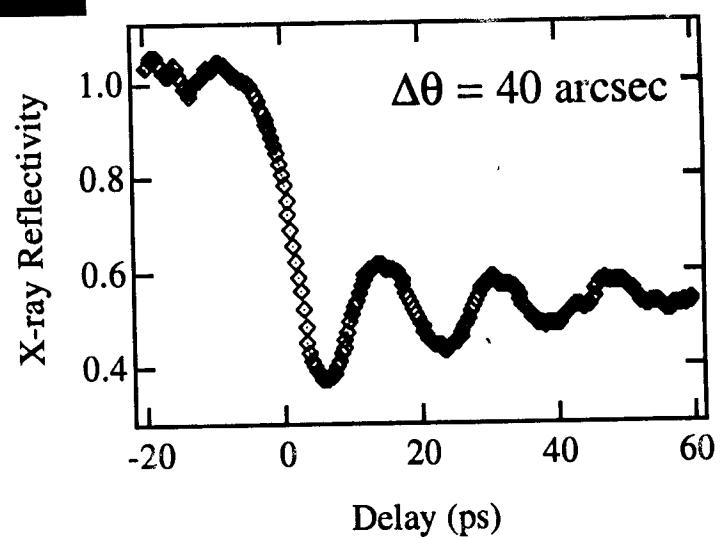
Streak Images of
Unperturbed (top)
and Laser-perturbed
(middle) X-ray Pulses



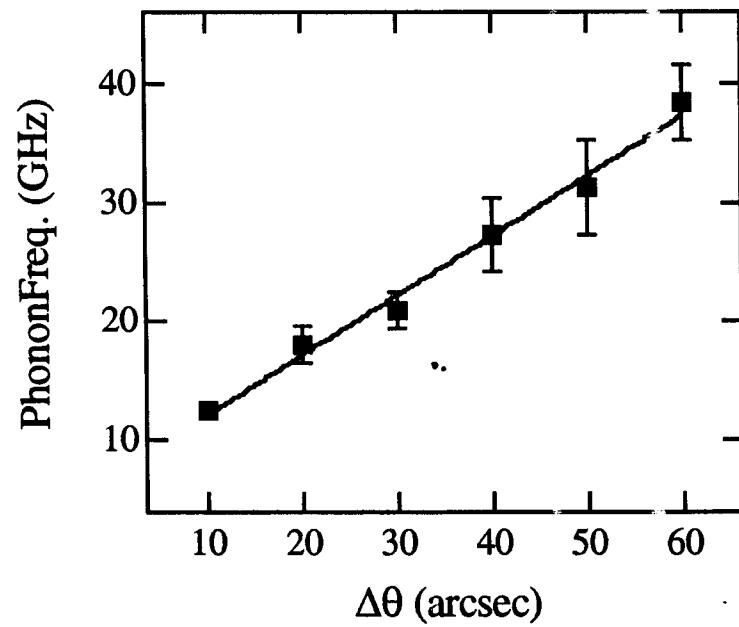
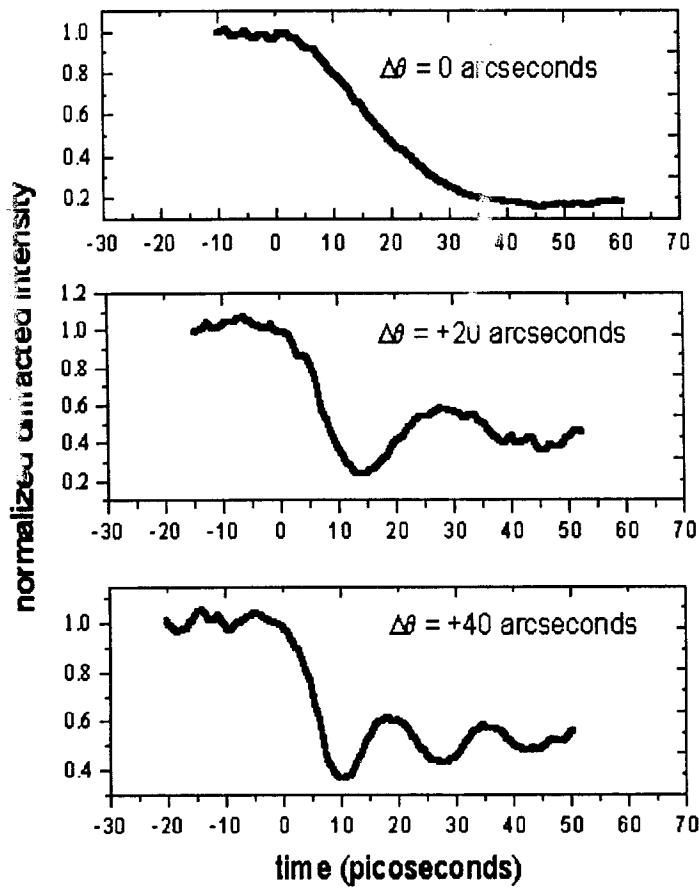
Advanced Light Source for Laser Materials Studies

X-ray Diffraction from
Laser Induced
Coherent Phonons (bottom)

Dynamical X-ray Scattering from Warm Matter



Spectroscopy of coherent acoustic phonons

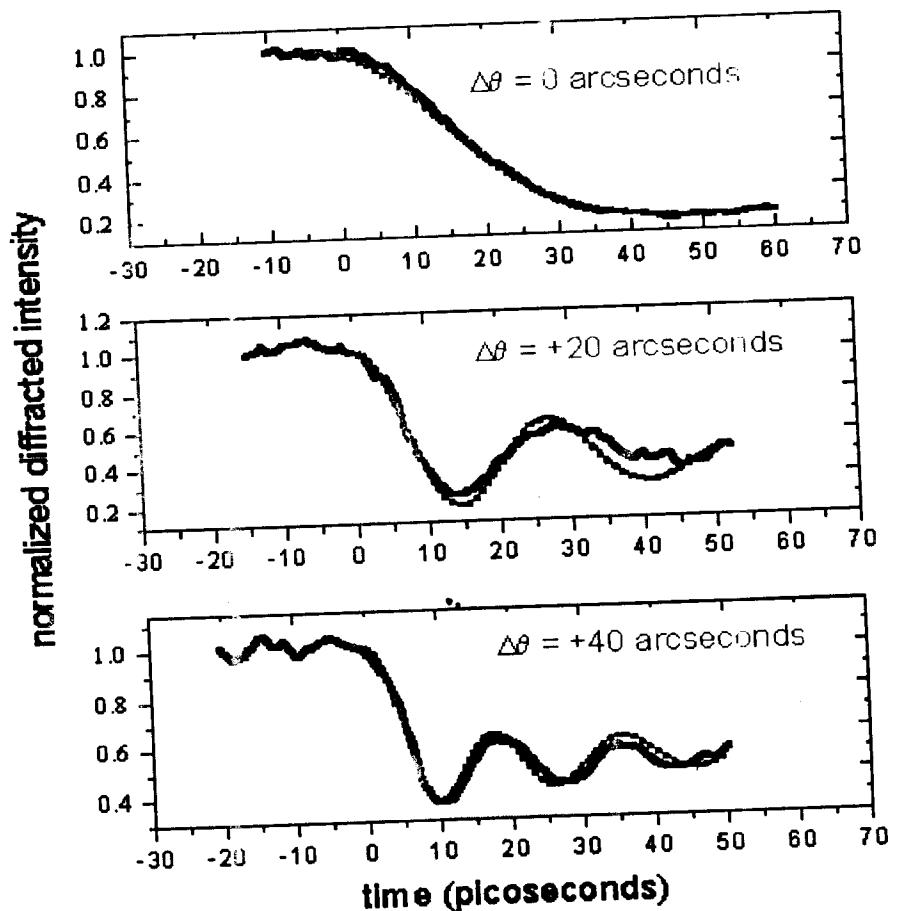


slope $\sim v_L \sim 4 \text{ km/s}$ (speed of sound)

Modeling the data

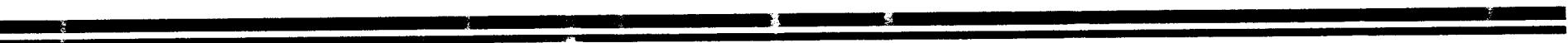
Fitting parameters:

- thermal strain = 0.17%
- non-thermal strain = 0.08%
- thermal electron-acoustic phonon coupling time = 12 ps

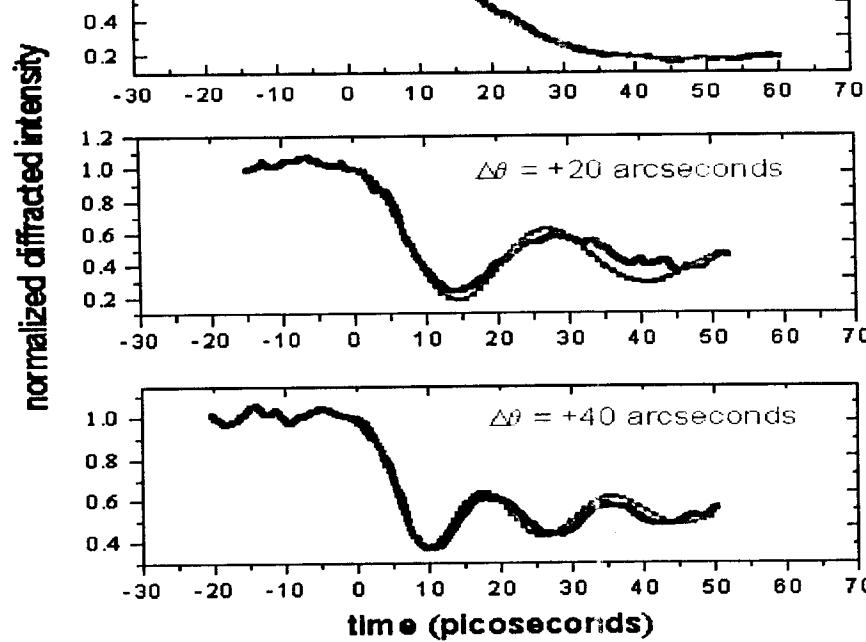


We observe coherent acoustic oscillations with amplitude corresponding to the amplitude expected from incoherent phonons at the melting temperature

Temporal behavior depends critically on laser fluence

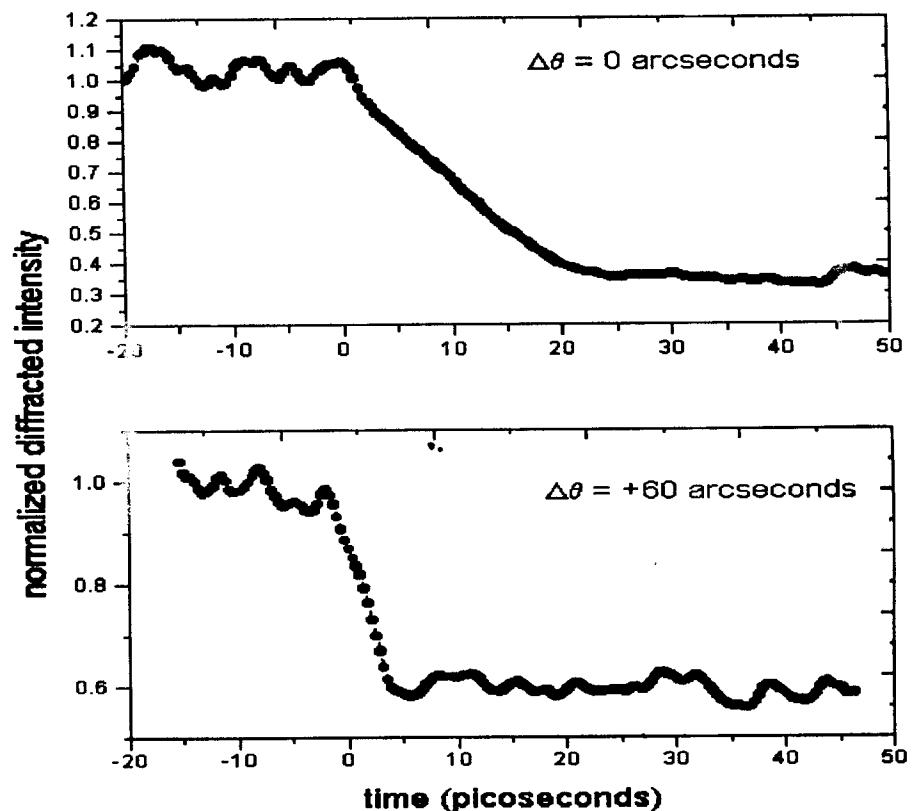


20 % below damage threshold



10 % below damage threshold

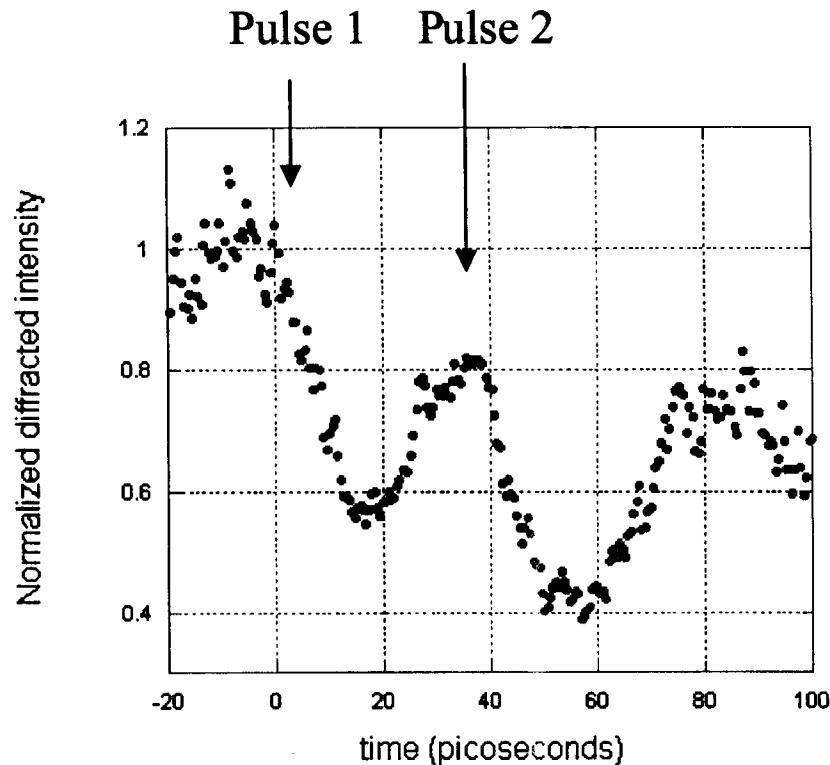
~60,000 shots



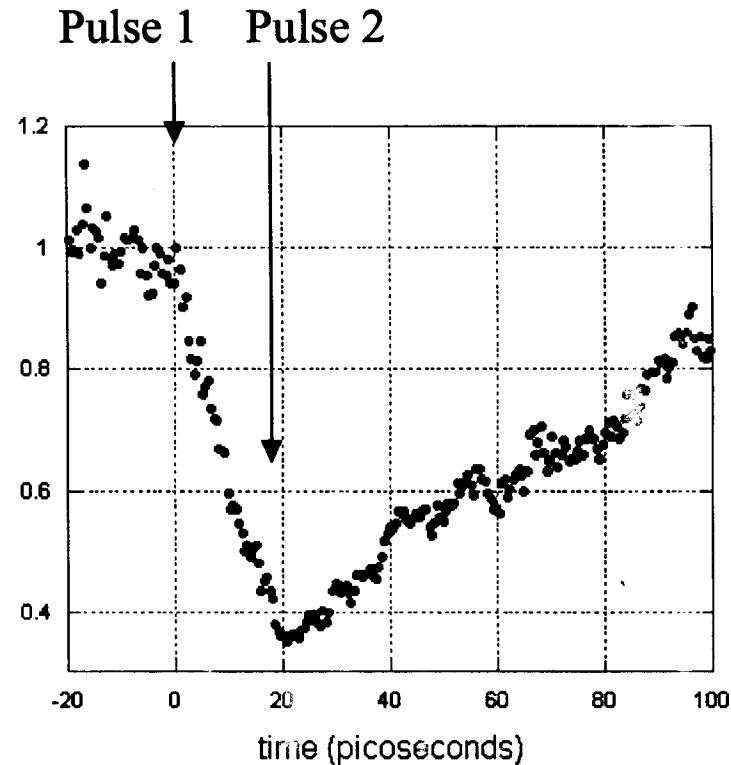
Coherent control of acoustic phonons in bulk InSb

- Control of x-ray diffraction from crystals
- The creation of highly non-equilibrium states of matter
- Investigation of nonlinear acoustical effects

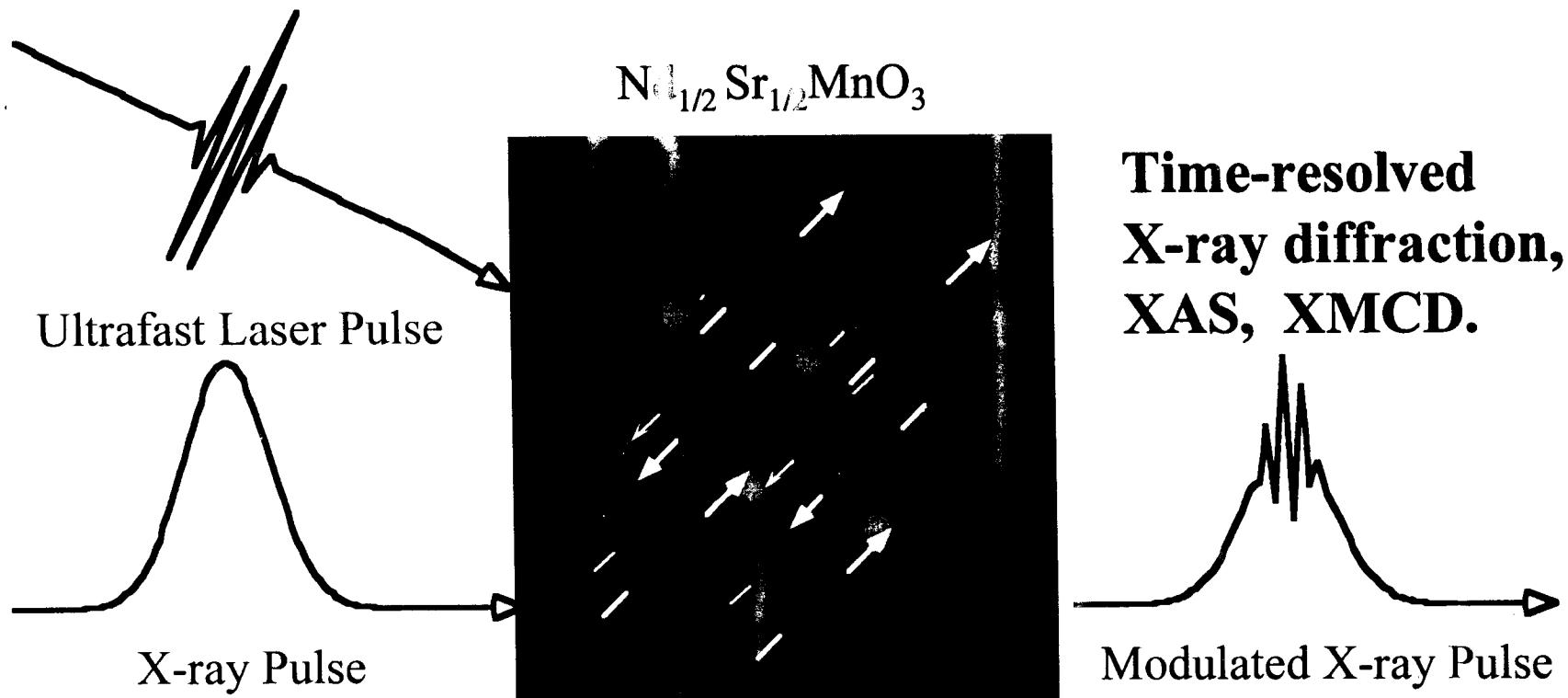
Constructive Interference
(2 pulses, 35 ps apart)



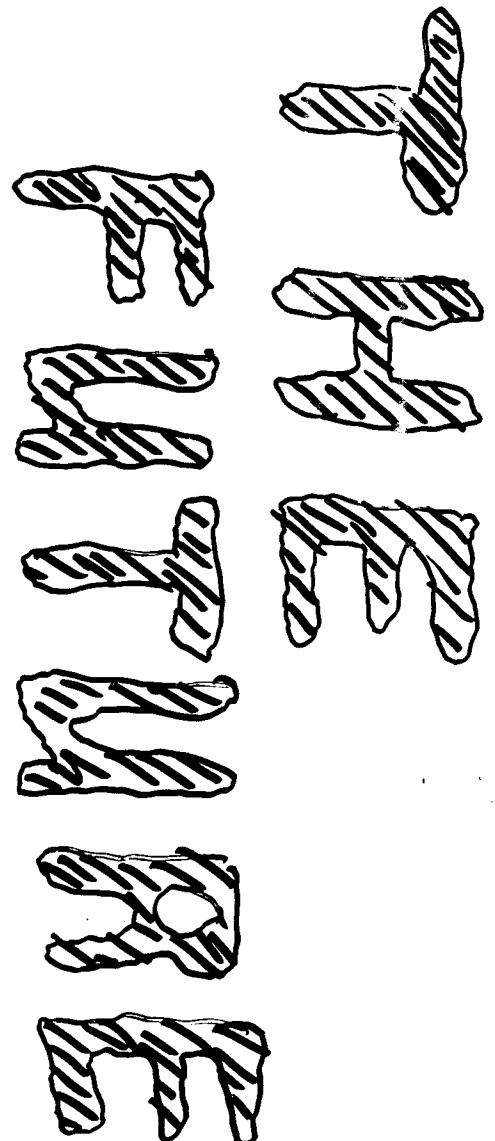
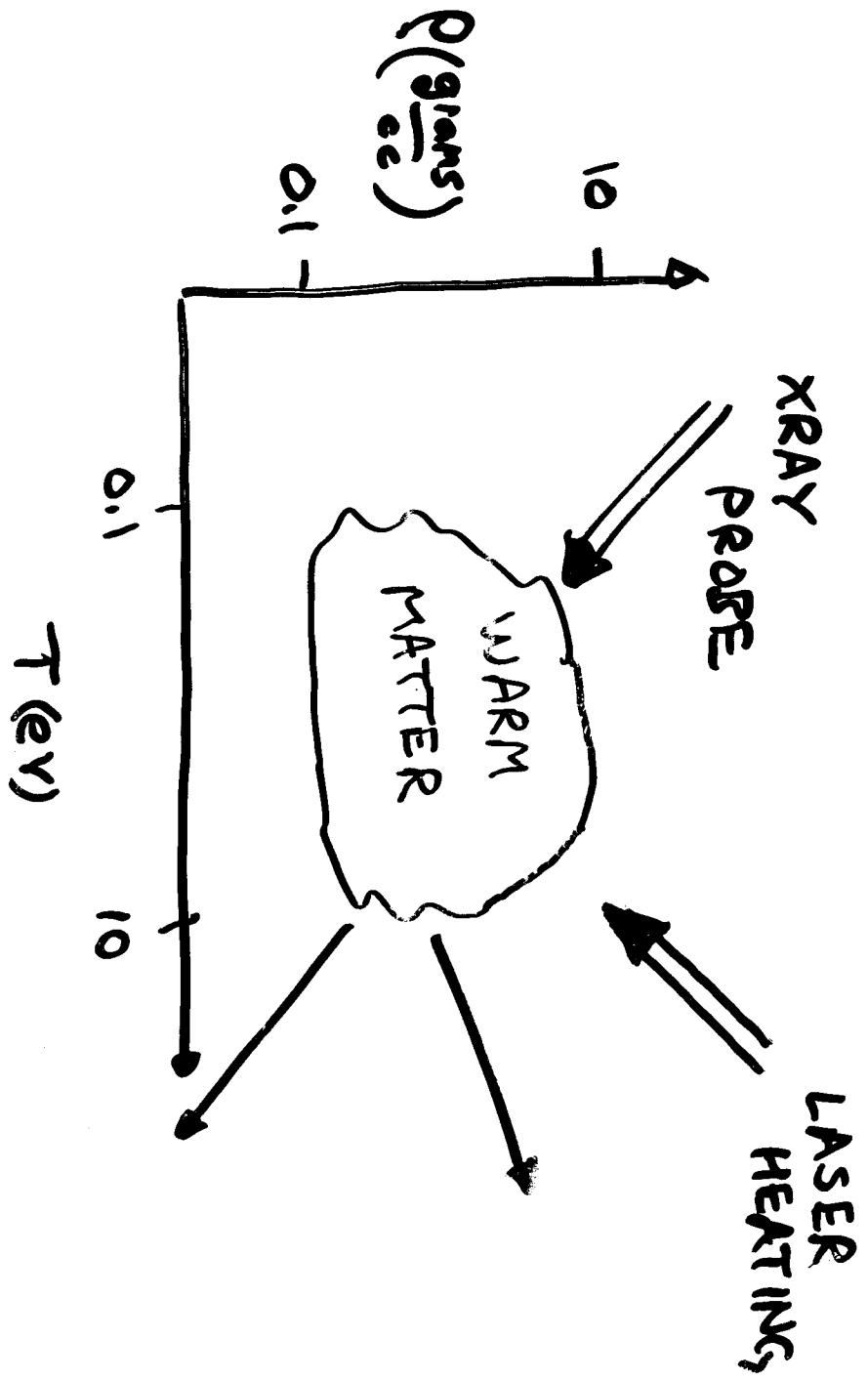
Destructive Interference
(2 pulses, 18 ps apart)



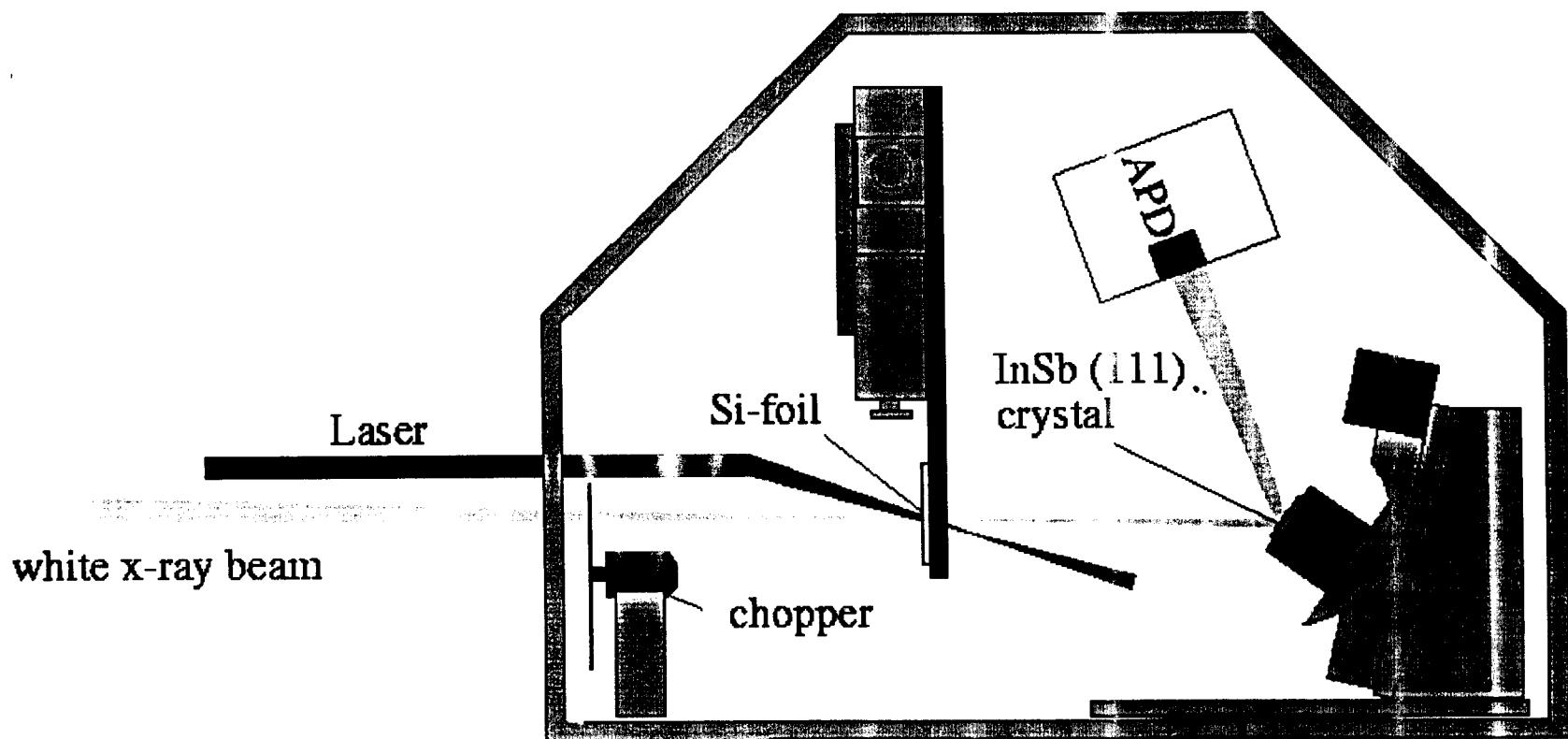
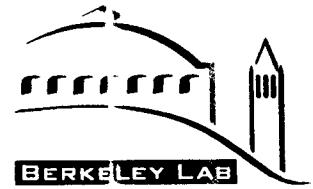
Time-Resolved X-ray Measurements of Polaron Dynamics of Charge-Ordered $\text{Nd}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$

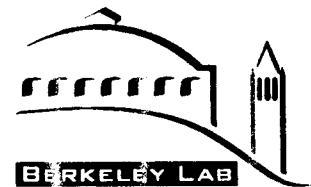


- Ultrafast laser pulses induce charge-transfer transition.
- Monitor lattice and spin dynamics with time-resolved x-ray measurements

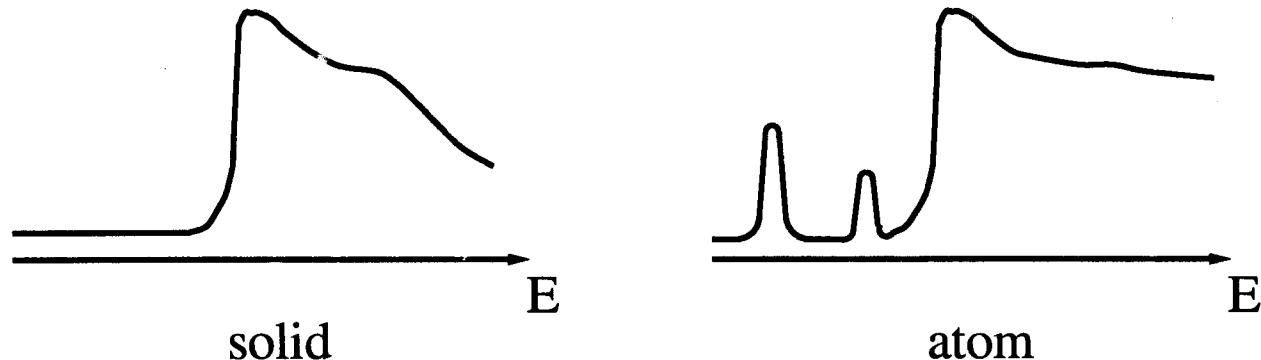


Si photoabsorption schematic diagram





Photoabsorption of laser excited foils

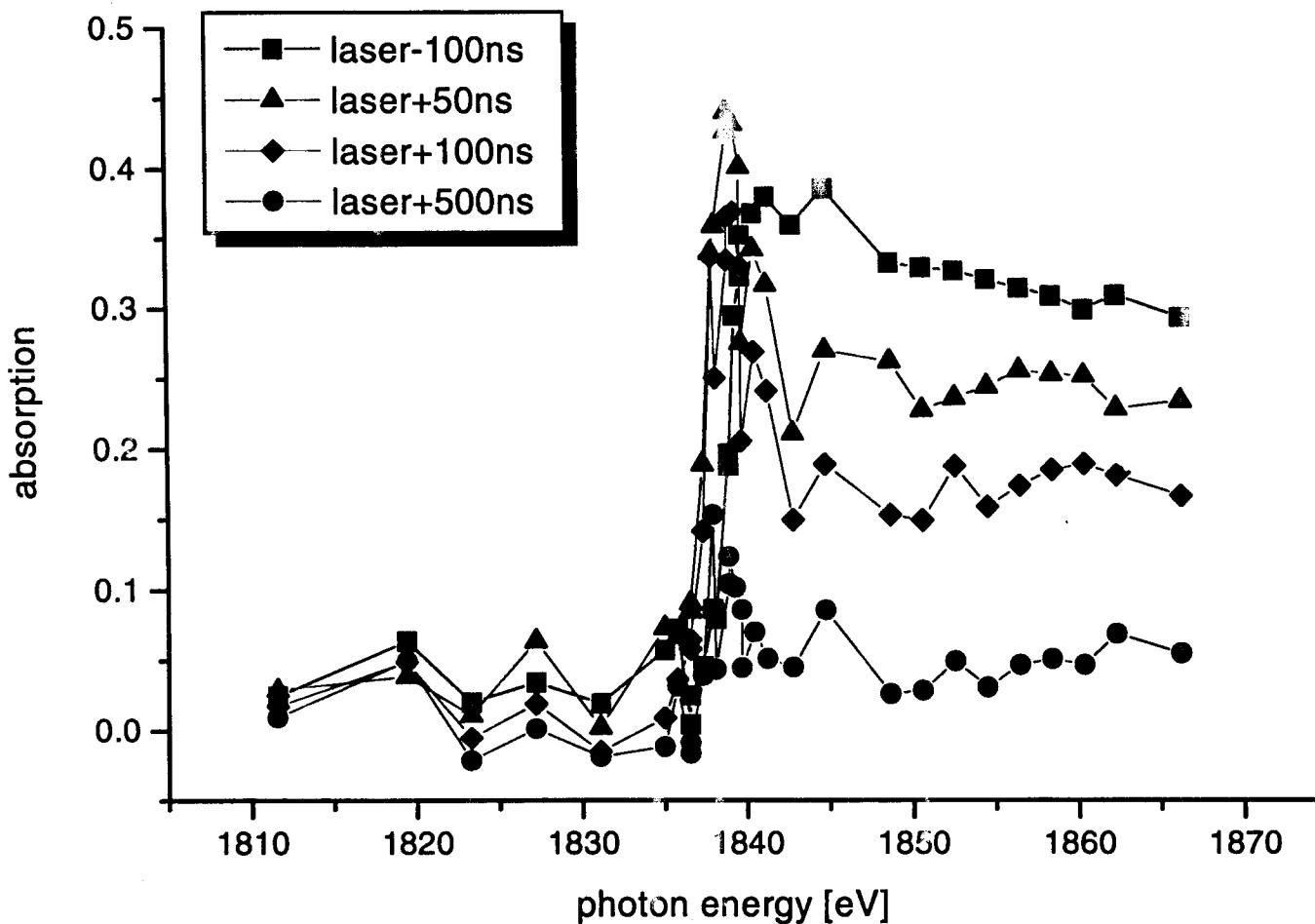


- Near-edge absorption (NEXAFS) as a probe for the change of electronic structure with the transitions from solid to liquid to gas.
- EXAFS as a probe of structure: nearest neighbor distances, coordination numbers
- Heat thin ($0.5 \mu\text{m}$) Si foil with ns pulse from Nd : YAG laser. Create uniform temperature and density.
- Earlier calculations and experiments:
 - Liquid Si is metallic, has 6.5 nearest neighbors (Gaspard et al., Phil. Mag. B 1984)
 - Pulsed laser irradiation of Si leads to clusters, liquid droplets and ionization (Murakami et al., PRL 1986)



NEXAFS of laser heated Si foil, higher fluence

fluence on Si-foil: 6.6 J/cm²



NEEDS

- STREAK CAMERA DETECTOR

- 100 fs resolution

- 100 ps range record length

- 1000:1 dynamic range

- 10% Q.E.

- 100 μm resolution

- AVALANCHE PHOTODIODE ARRAY

- "fast readout CCD with gain"

- 10 ns gate

- 100% Q.E.

- 300x gain

- 1-d 1000 pixels

- 2-d 1000 x 1000 pixels